Structure of Nanocrystalline Materials through Coherent X- ray Diffraction

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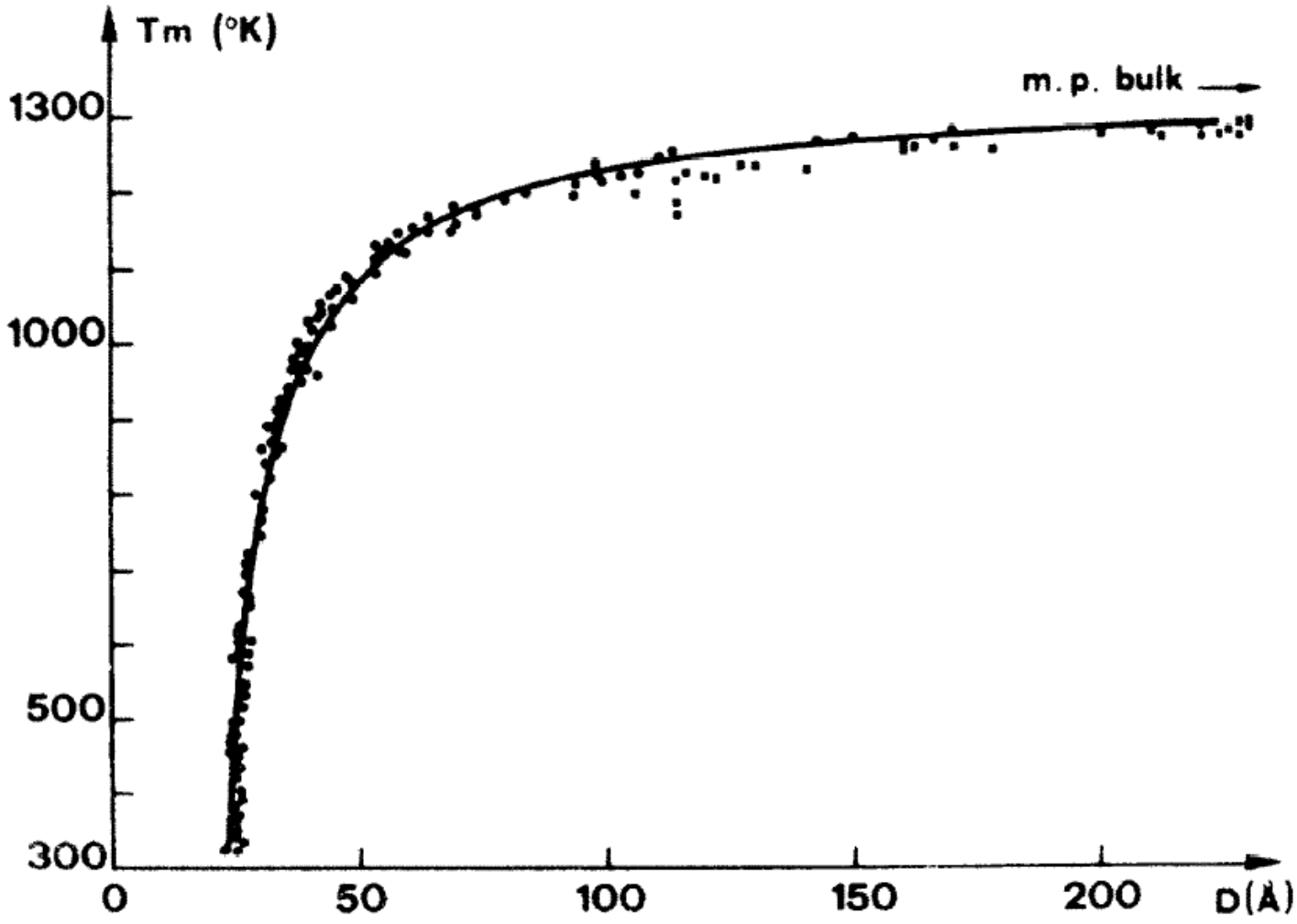
Outline

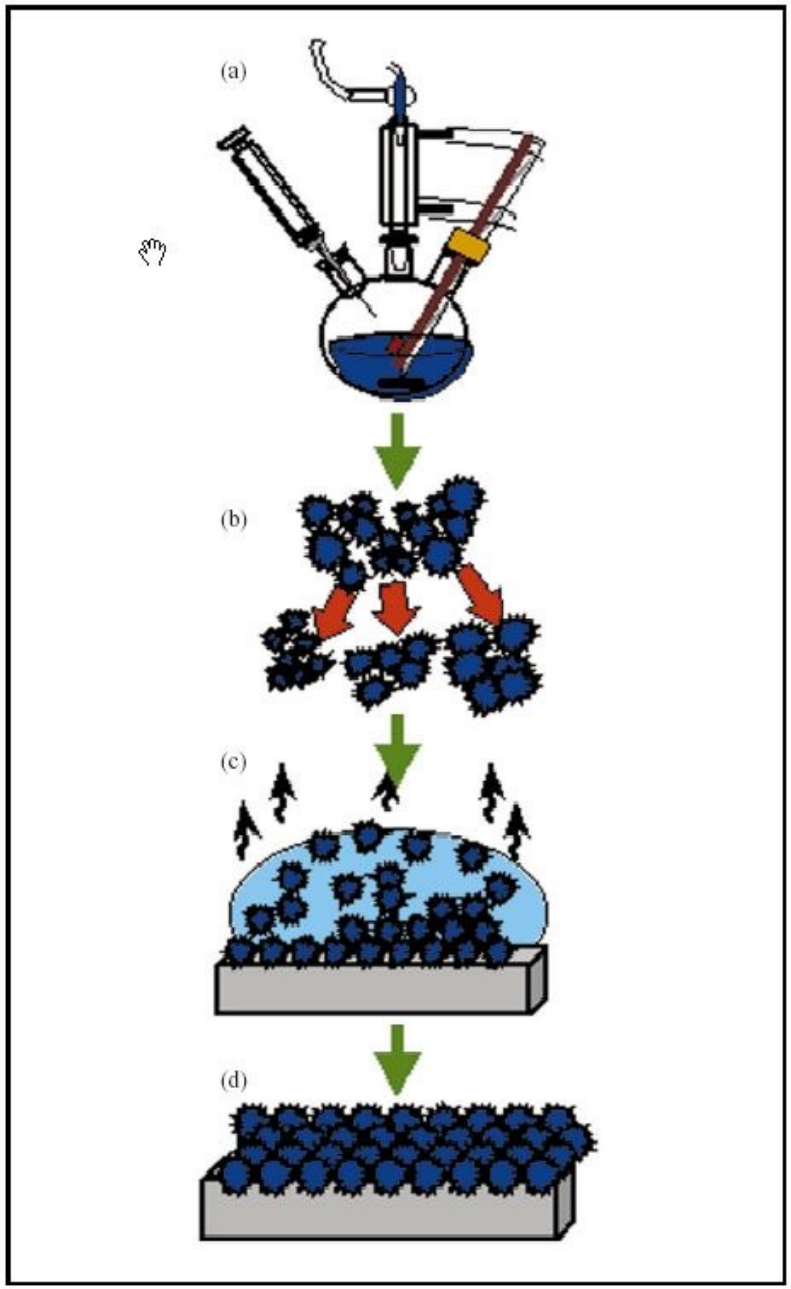
- Coherence in Diffraction
- The Phase Problem
- Nanocrystal Shapes
- Imaging of Phase Objects
- How small can we go?

Size-dependent Melting of Au Particles

P. Buffat and J-P. Borel, Phys. Rev. A 2287-97 (1975)

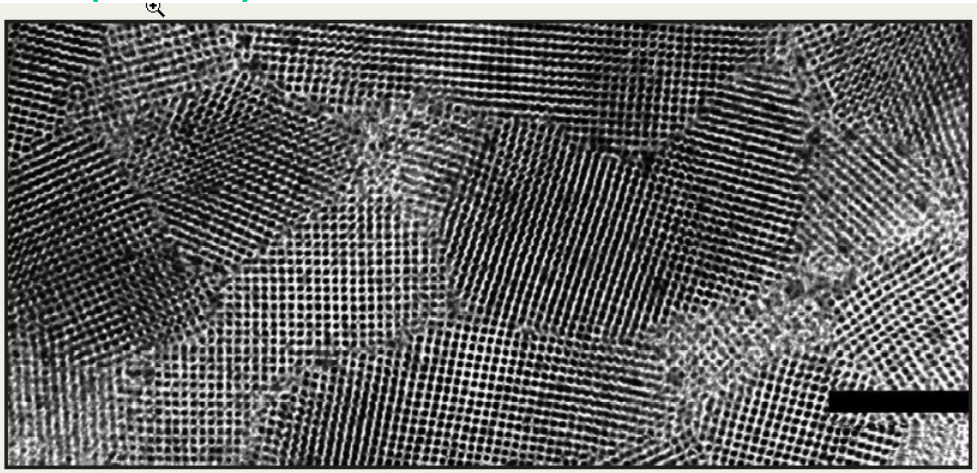


Chemical Synthesis of Nanocrystals

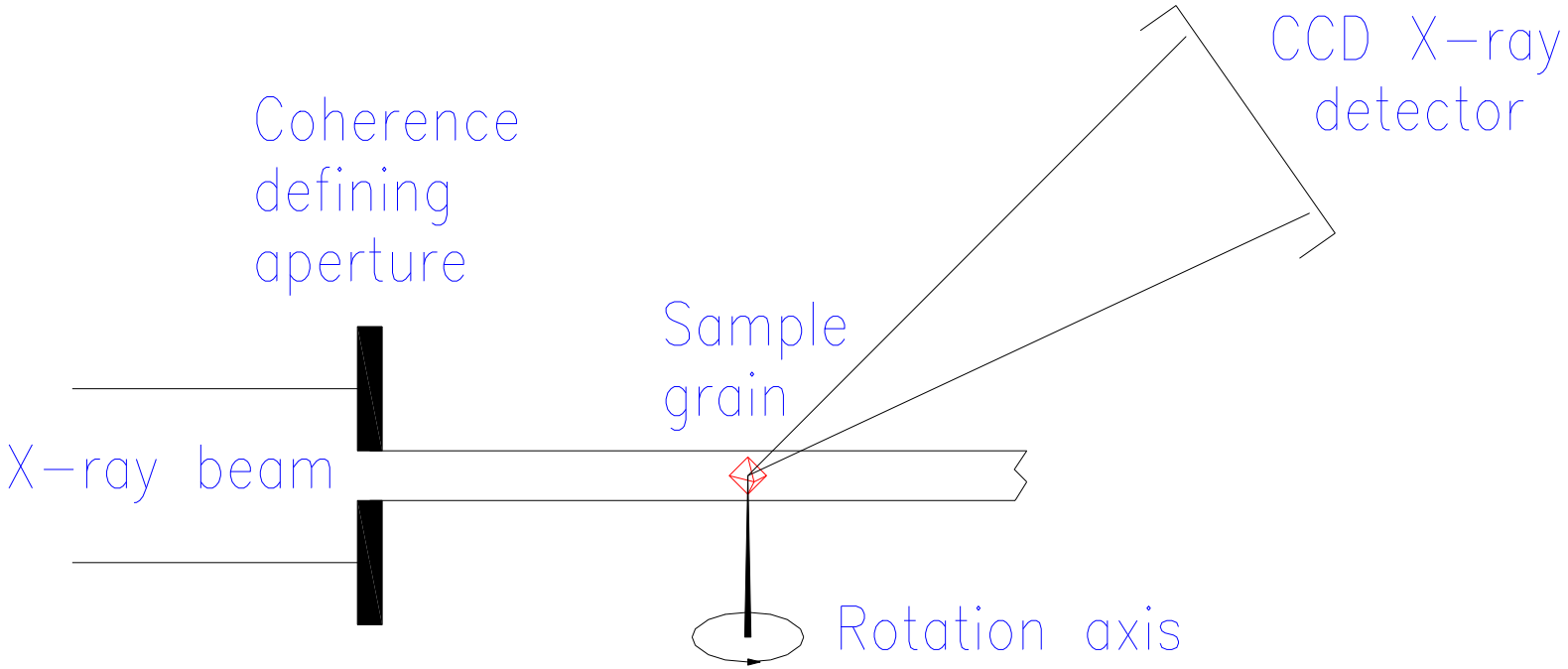


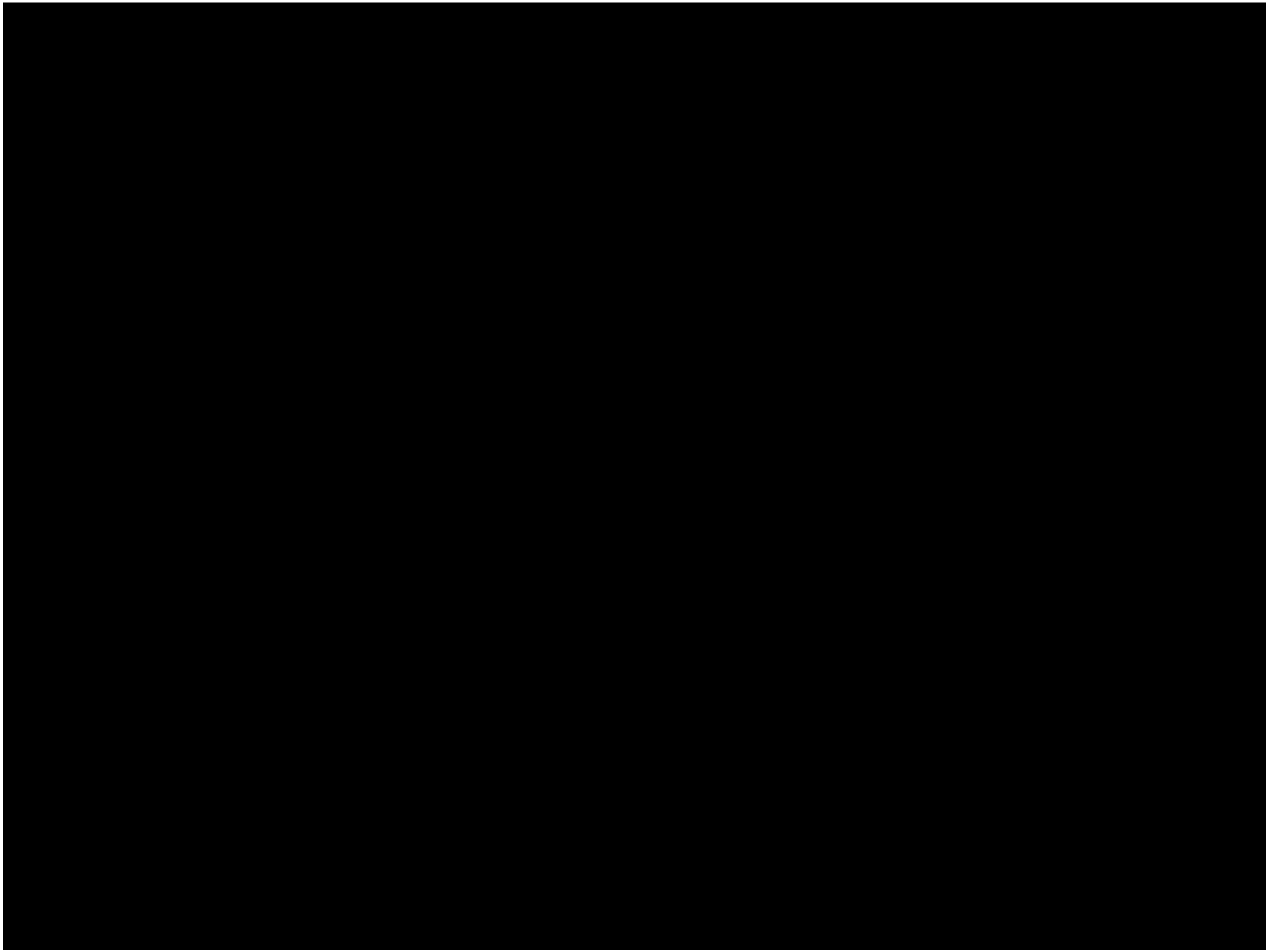
- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

C. B. Murray, IBM J. Res. & Dev. **45** 47 (2001)

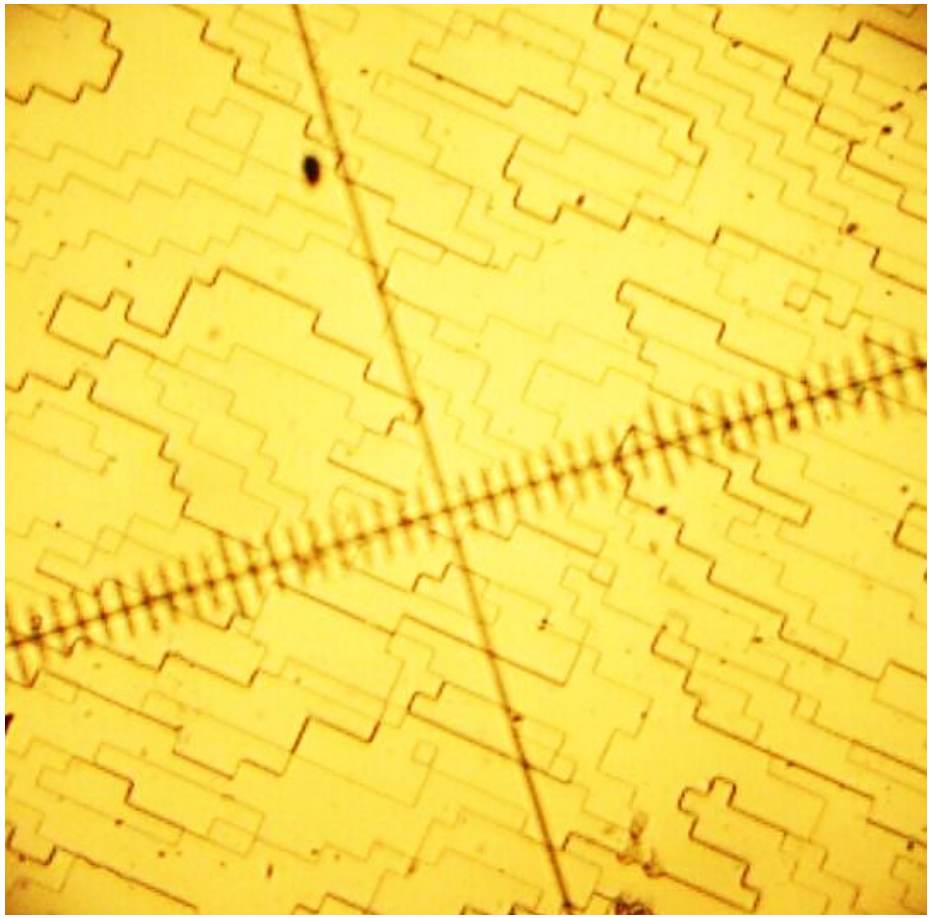


Lensless X-ray Microscope

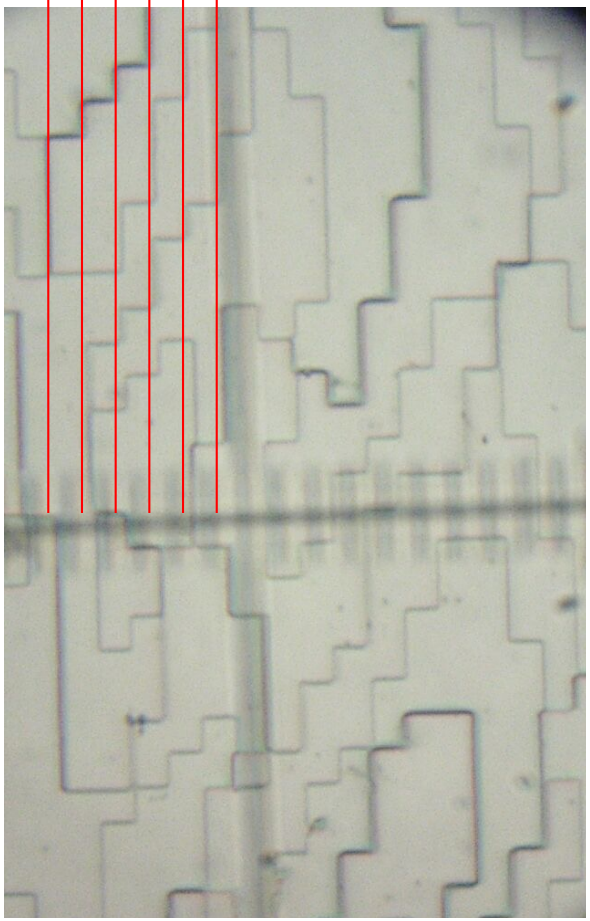




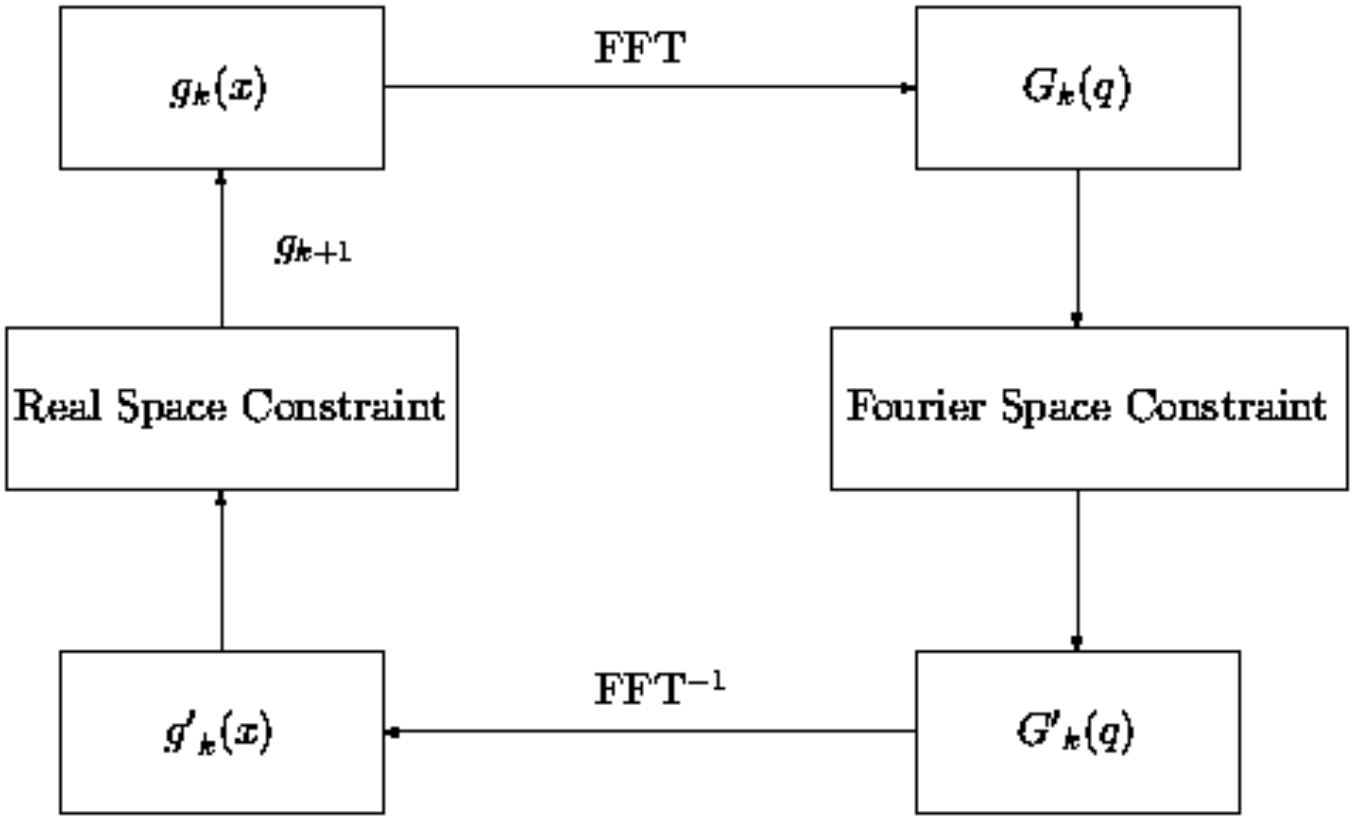
Microscope Images of Gratings



5μm grid lines



Generic "Error Reduction" method

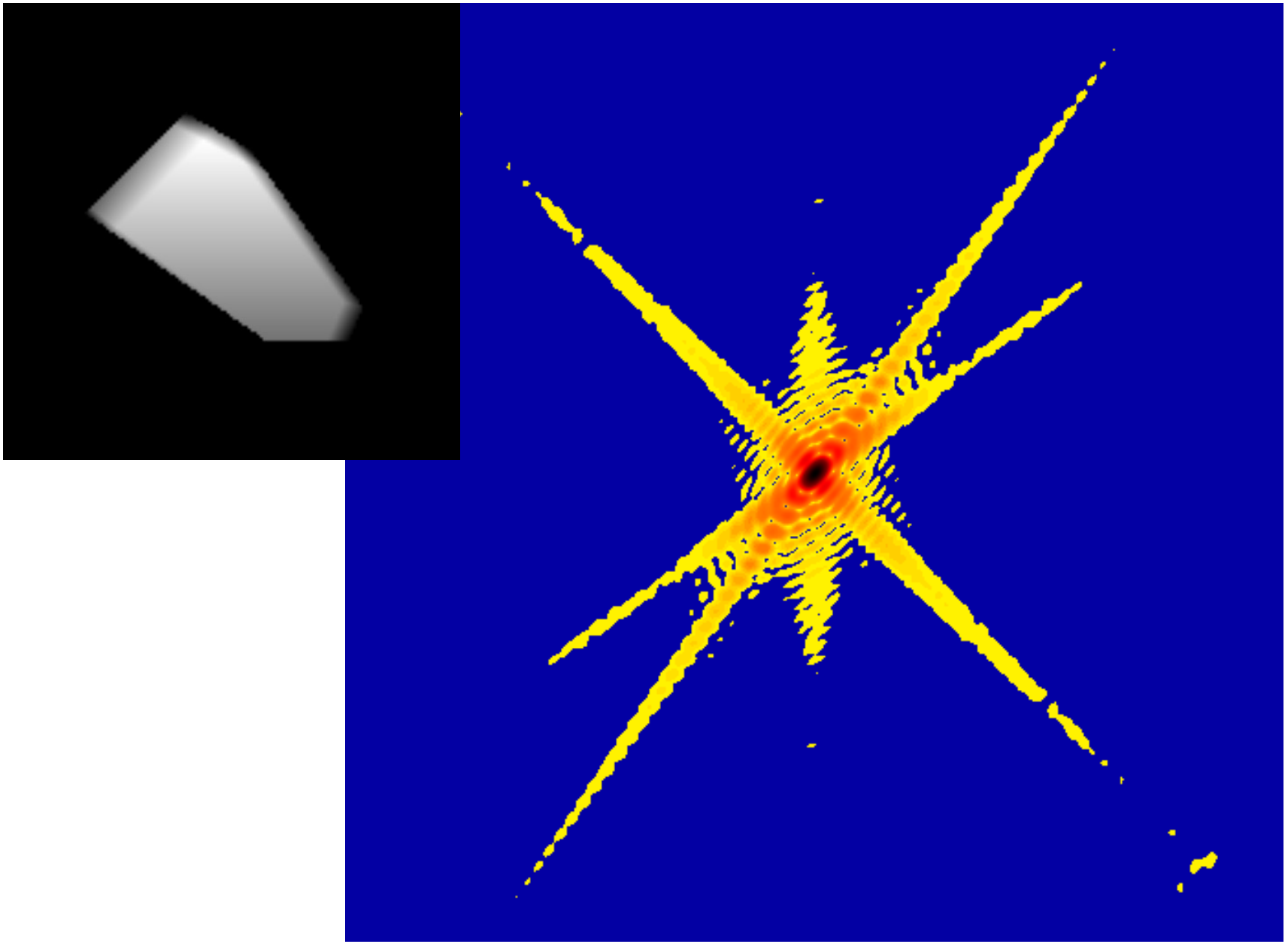


J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

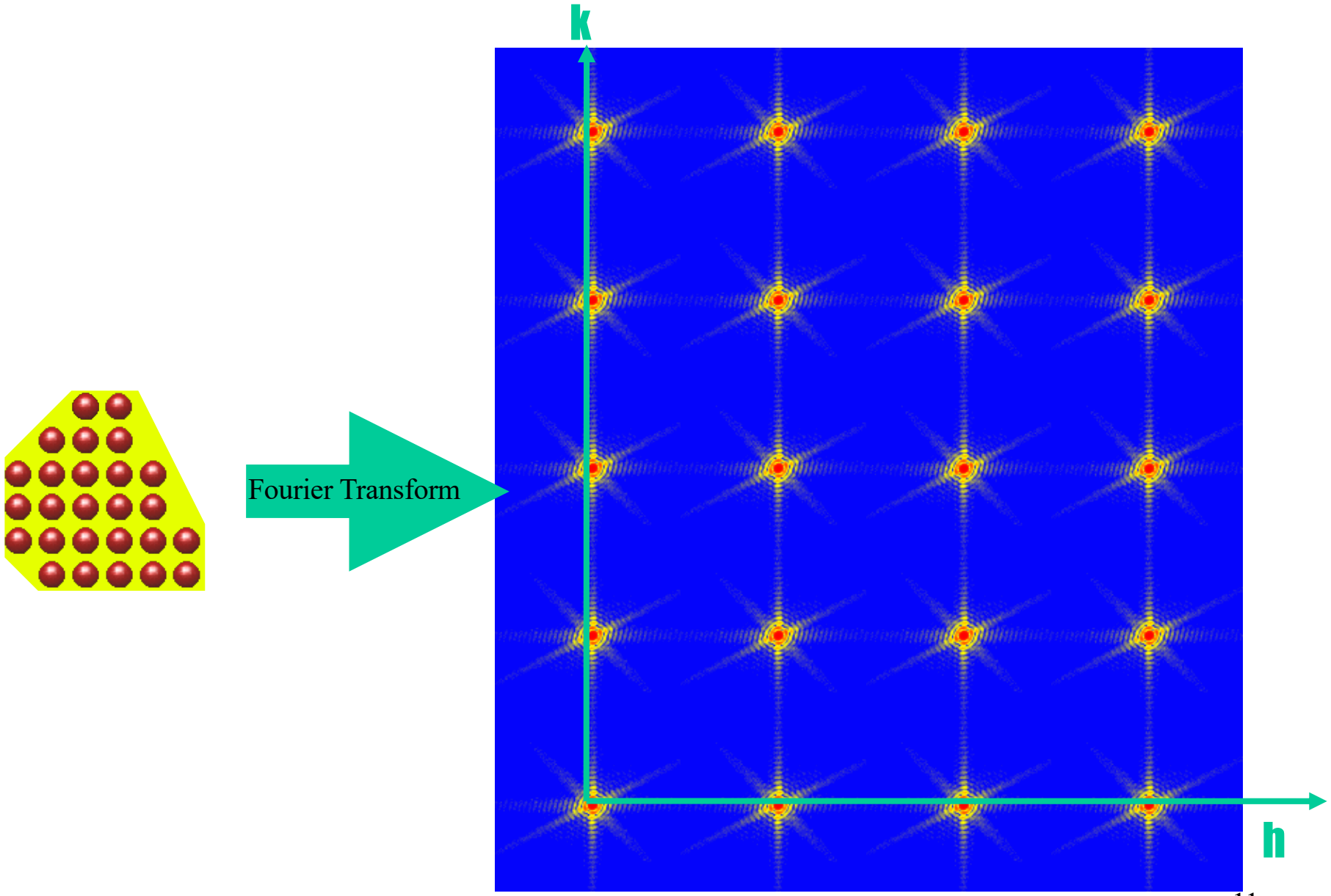
Real-space Constraints in Crystallograhy

R. P. Millane, J. Opt. Soc Am. A **13** 725 (1996)

- 'Positivity' and 'Atomicity' constraints (Sayre)
- Finite support, molecular envelope
- Solvent flattening/Molecular replacement
- Non-crystallographic symmetry
- Non-uniqueness is 'pathologically rare' (d>1)
- Uses memory to avoid stagnation (Fienup HIO)

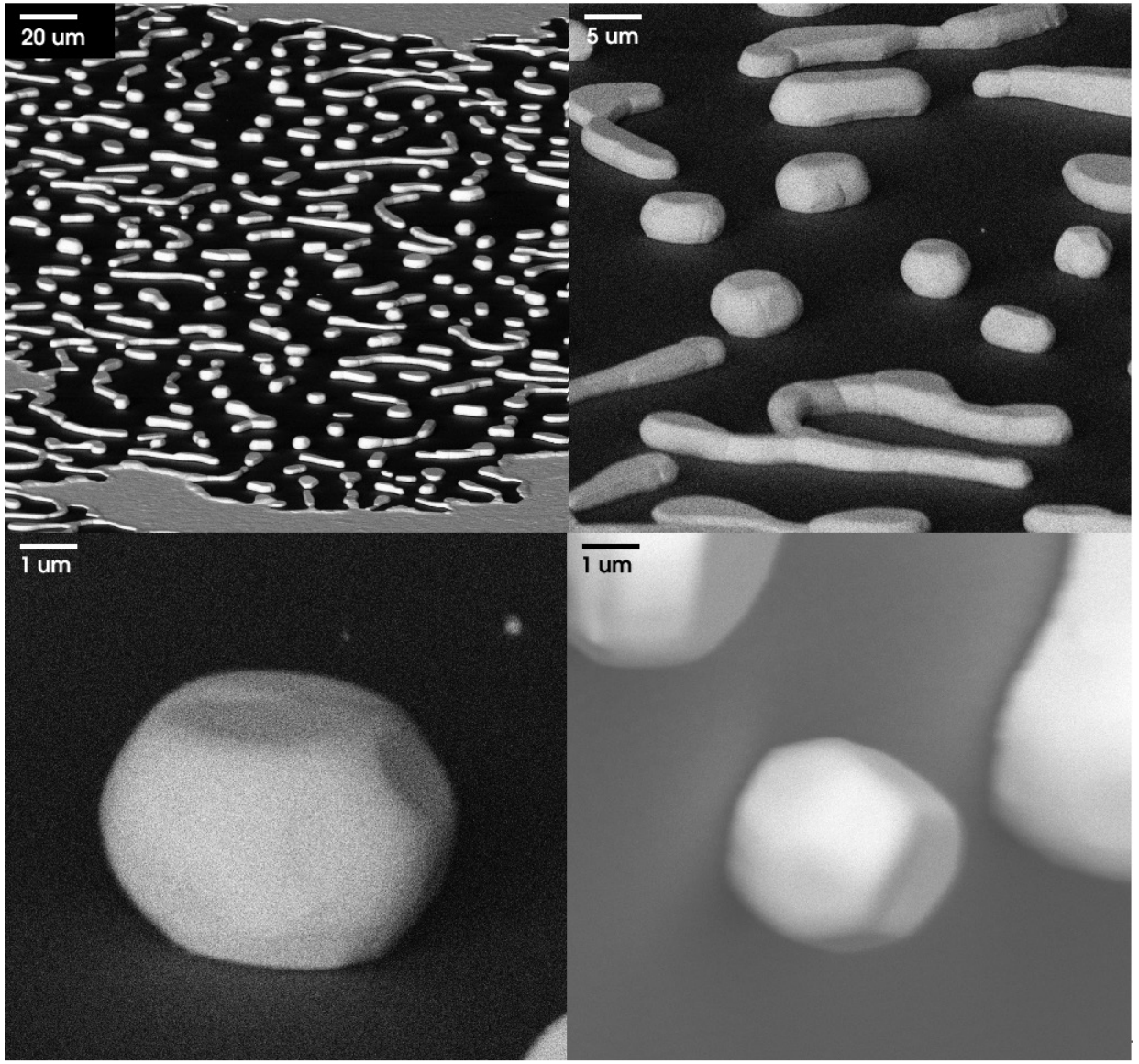


Coherent Diffraction from Crystals



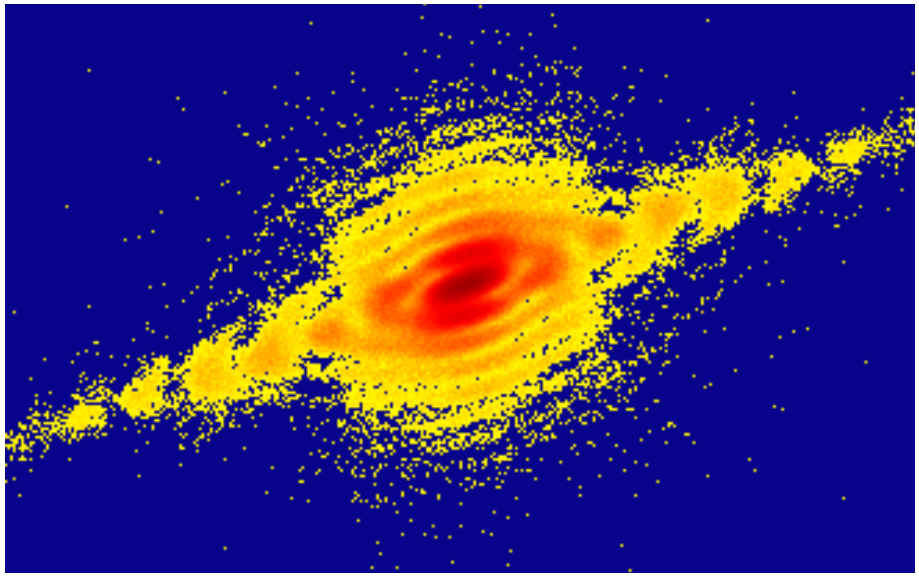
SEMS

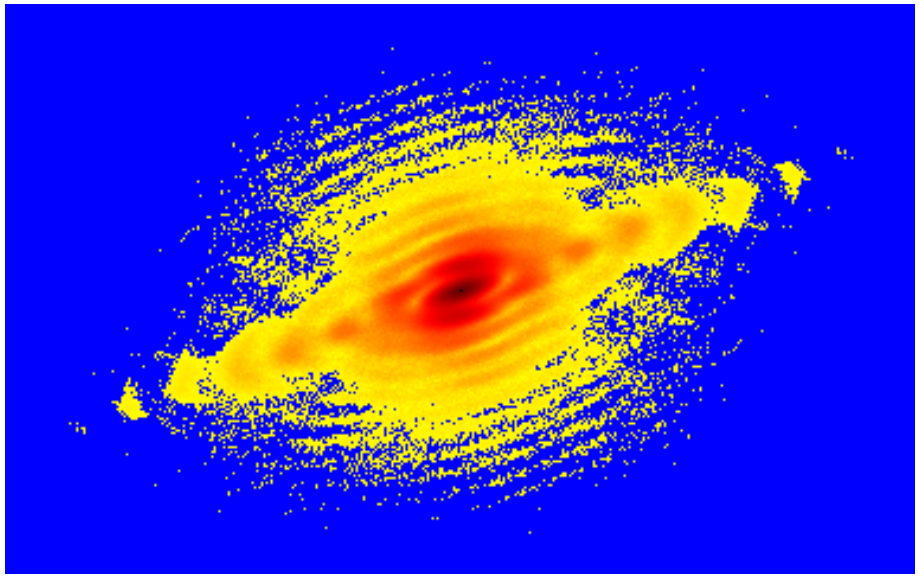
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.

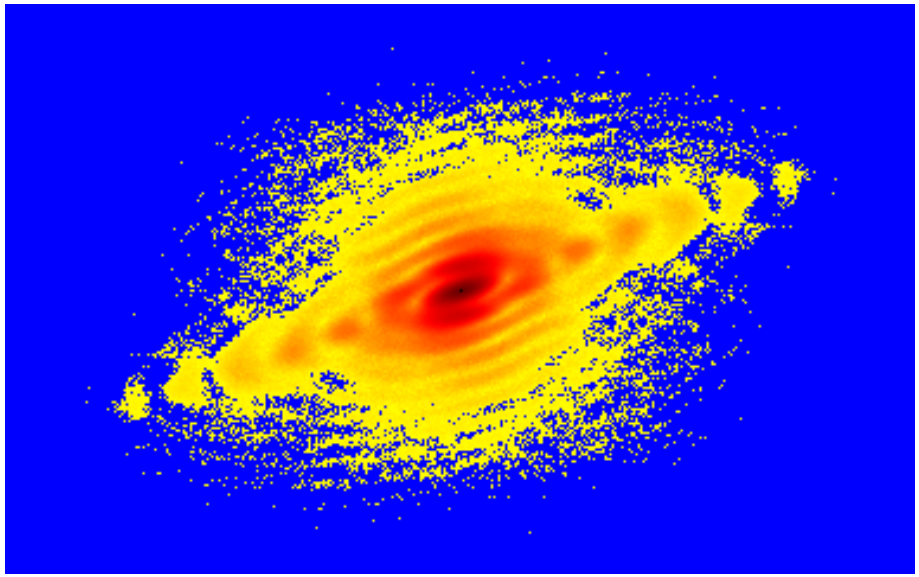


Symmetrized Data and two best fits

Chisq=0.0005

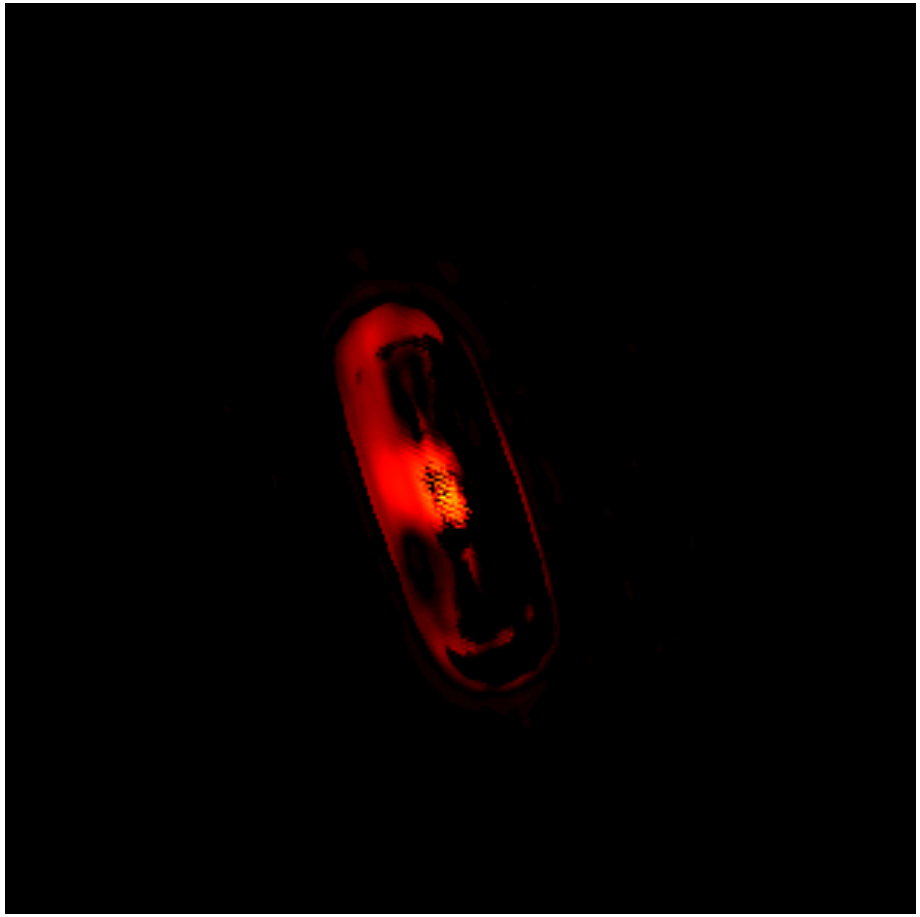


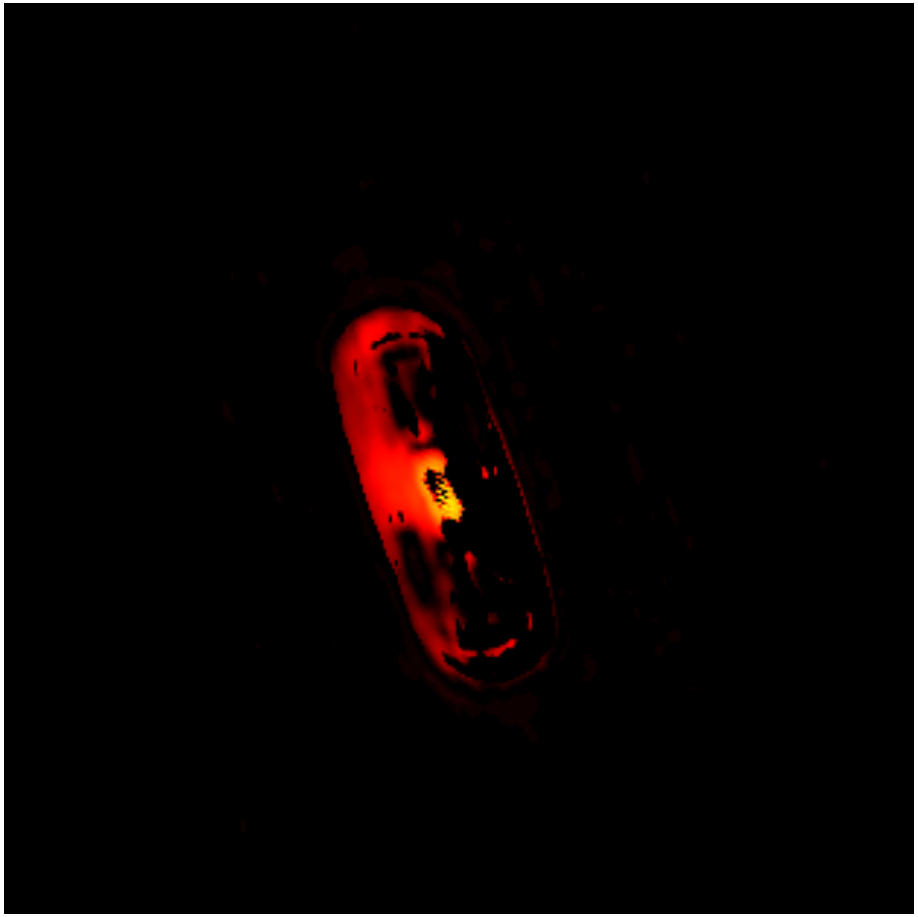




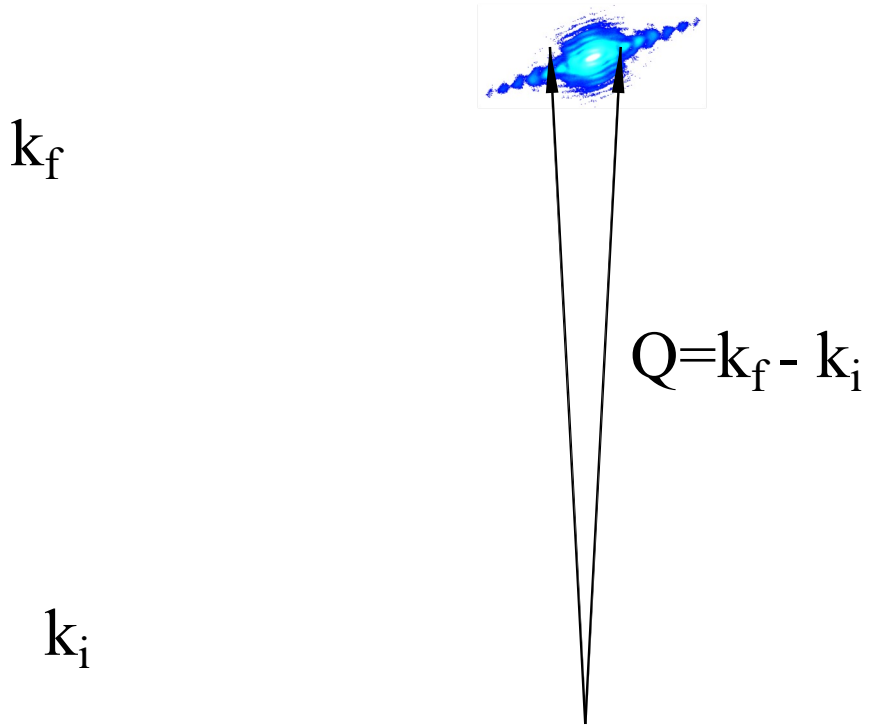
2D Reconstructions

chisquare = 0.0005

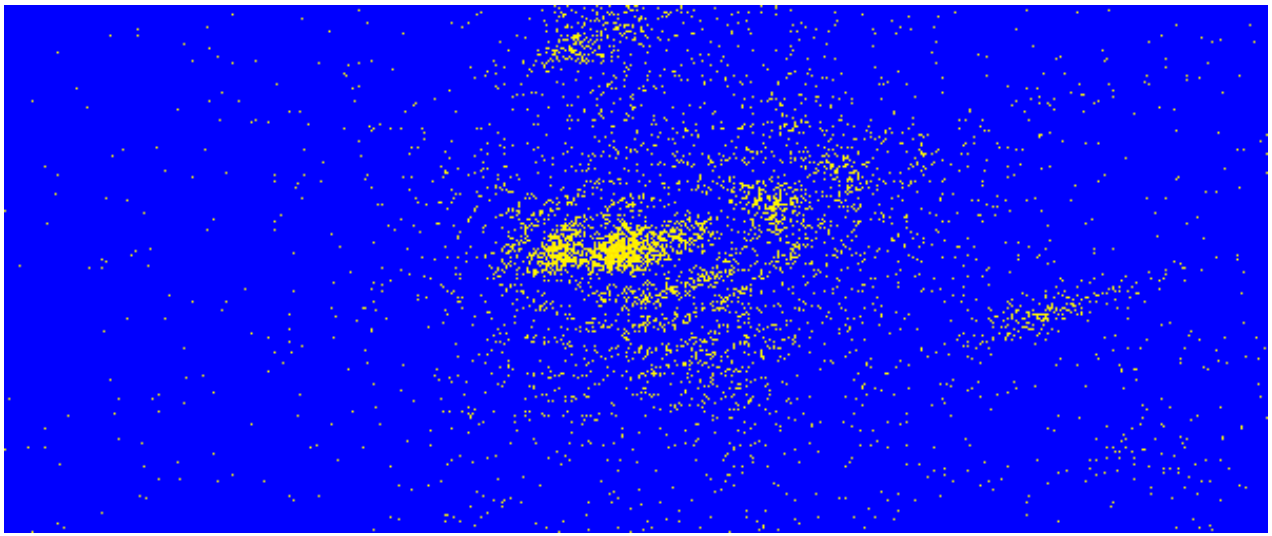




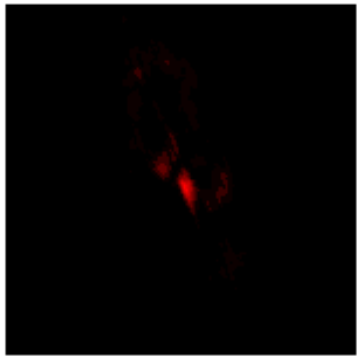
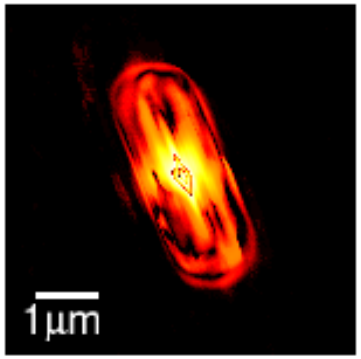
3D Diffraction Method

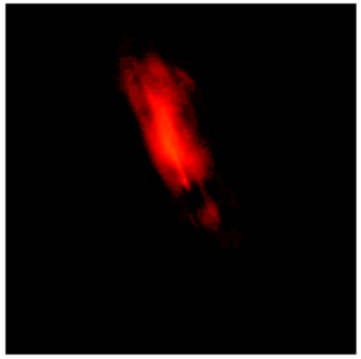
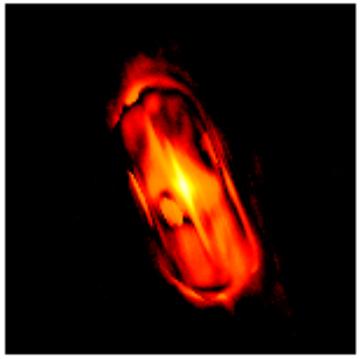
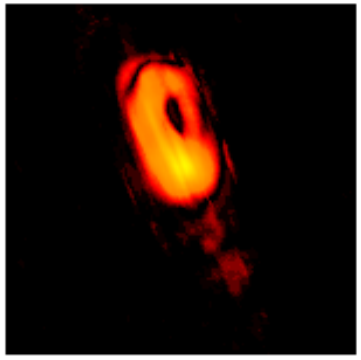
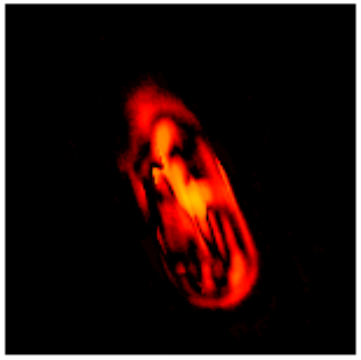
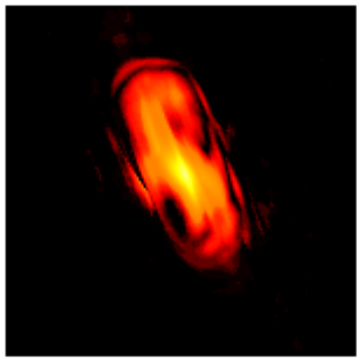
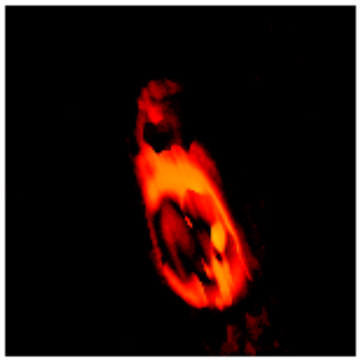
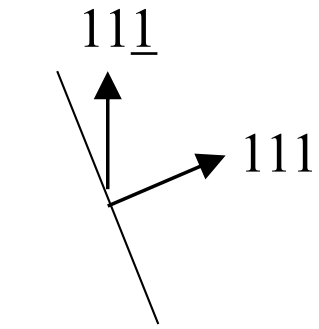


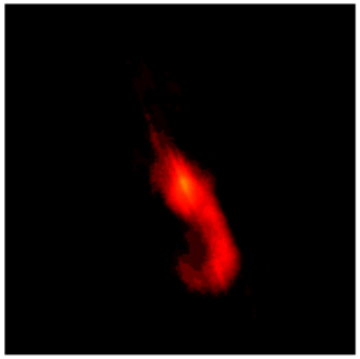
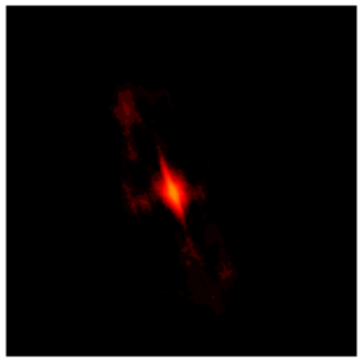
3D Diffraction Data
1 micron Au crystal



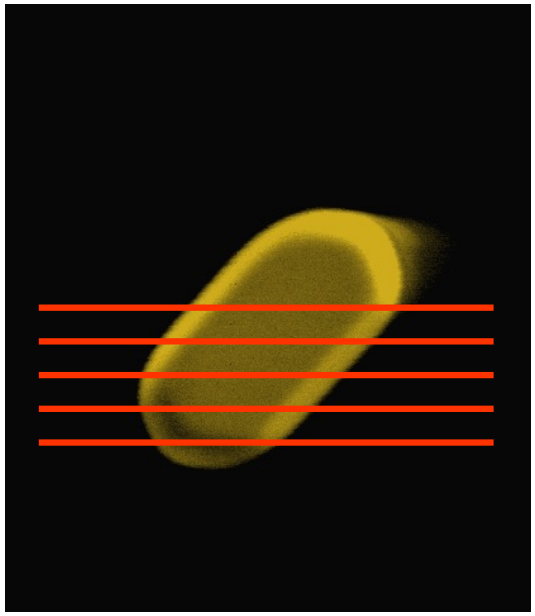
* Center is Symmetric *

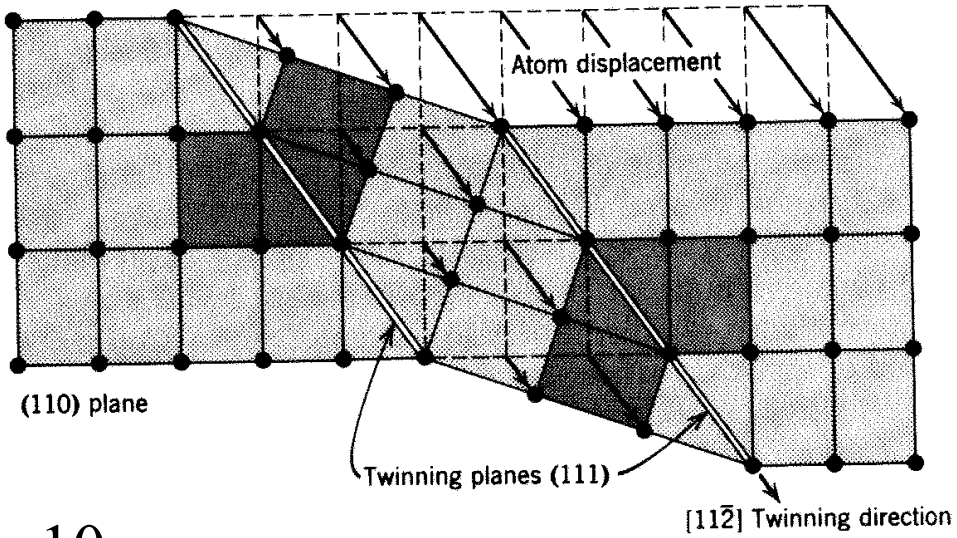



Slices through plan view SEM:



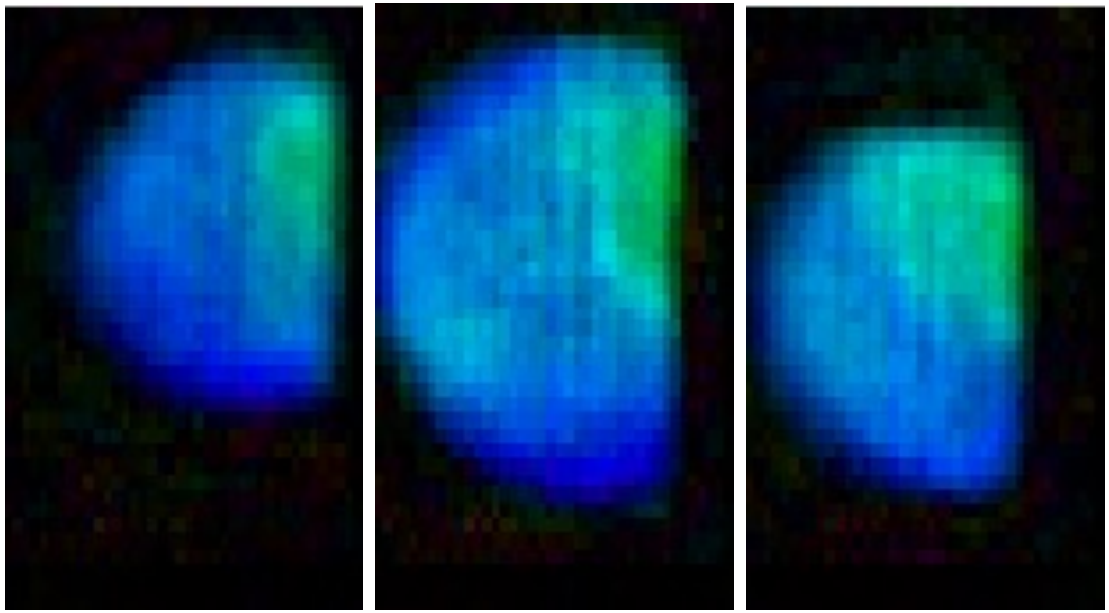
Twinning in deformed FCC metals

J. Wulff, "Structure and Property of Materials III" (1965)



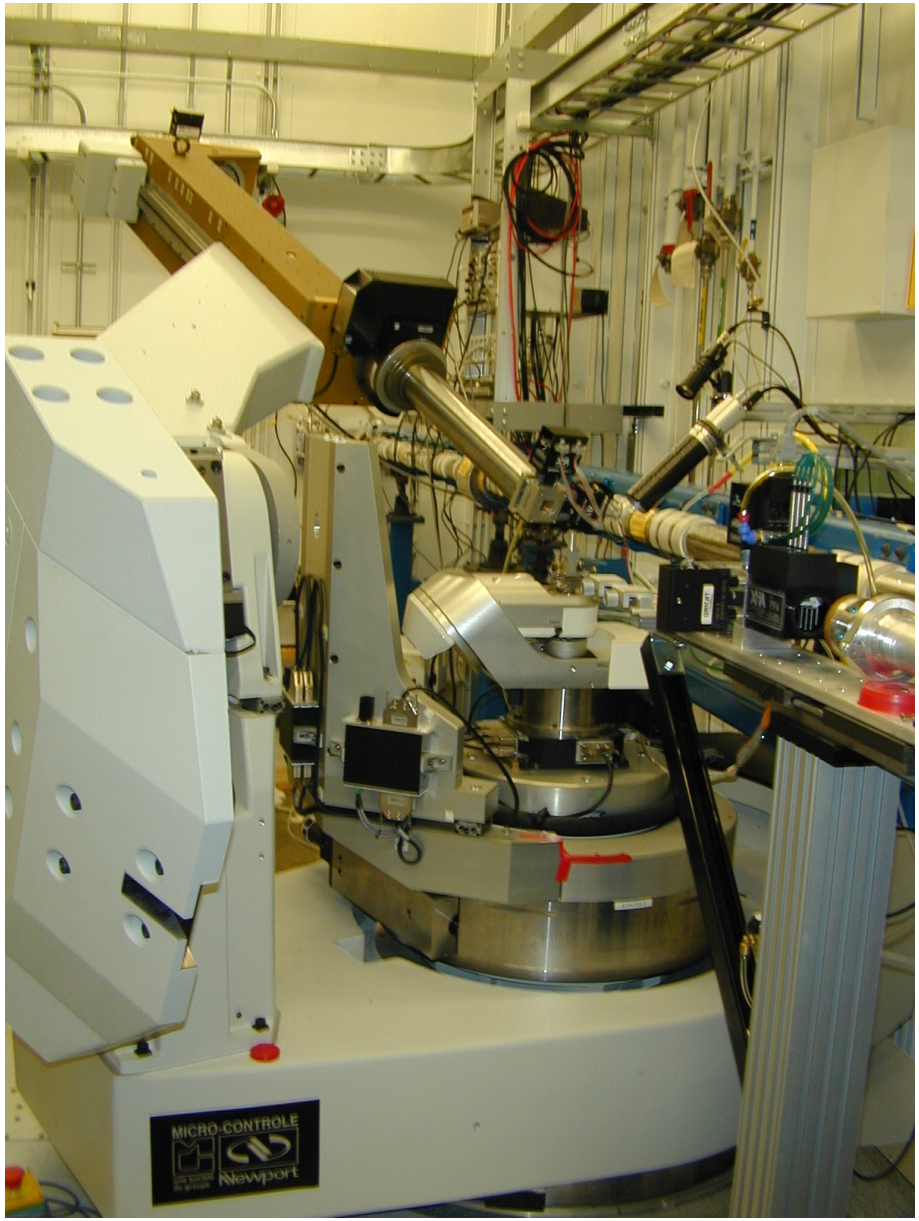
~10μm
Cu

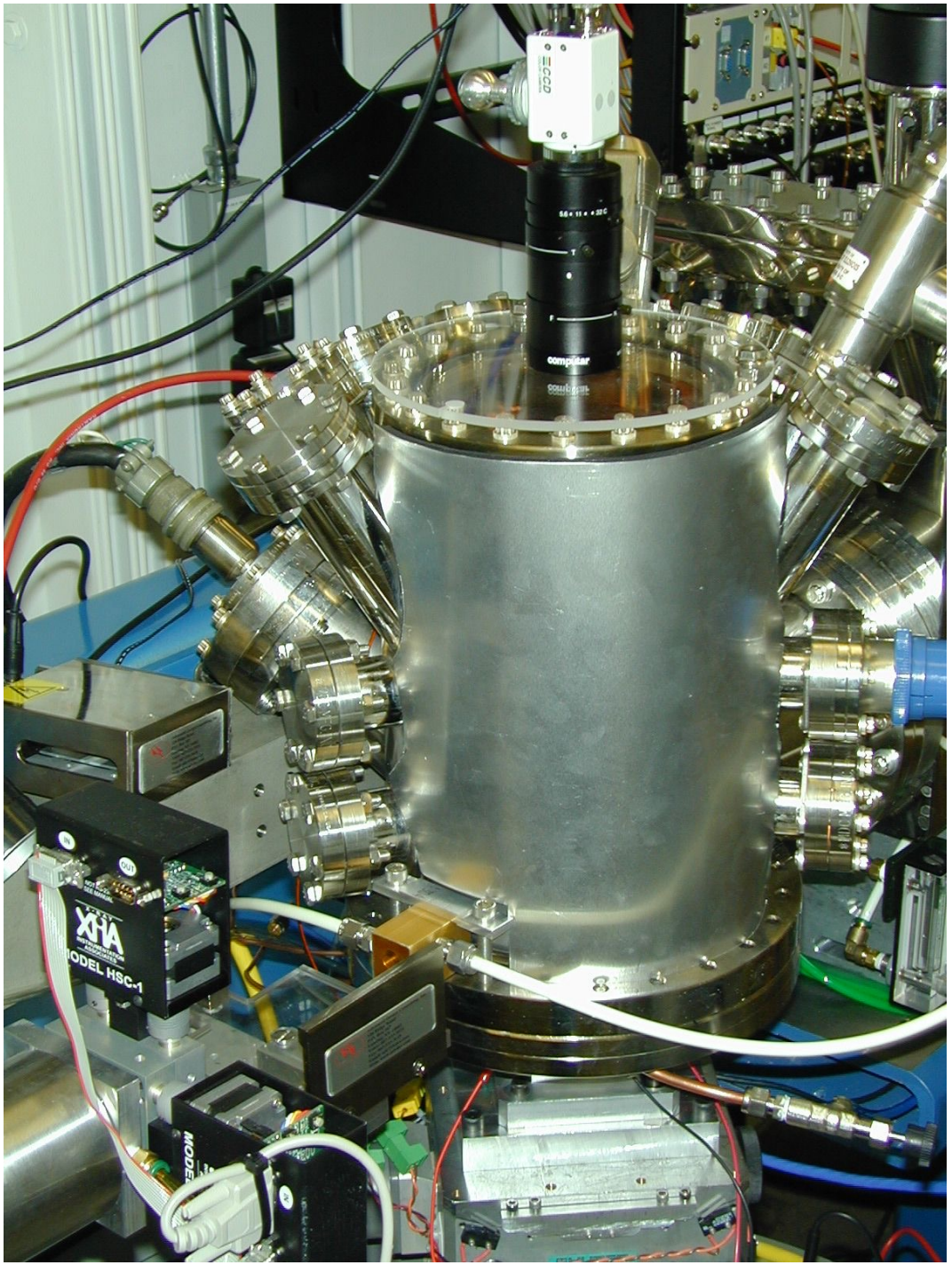
3D Phase Images of Lead Nanocrystals



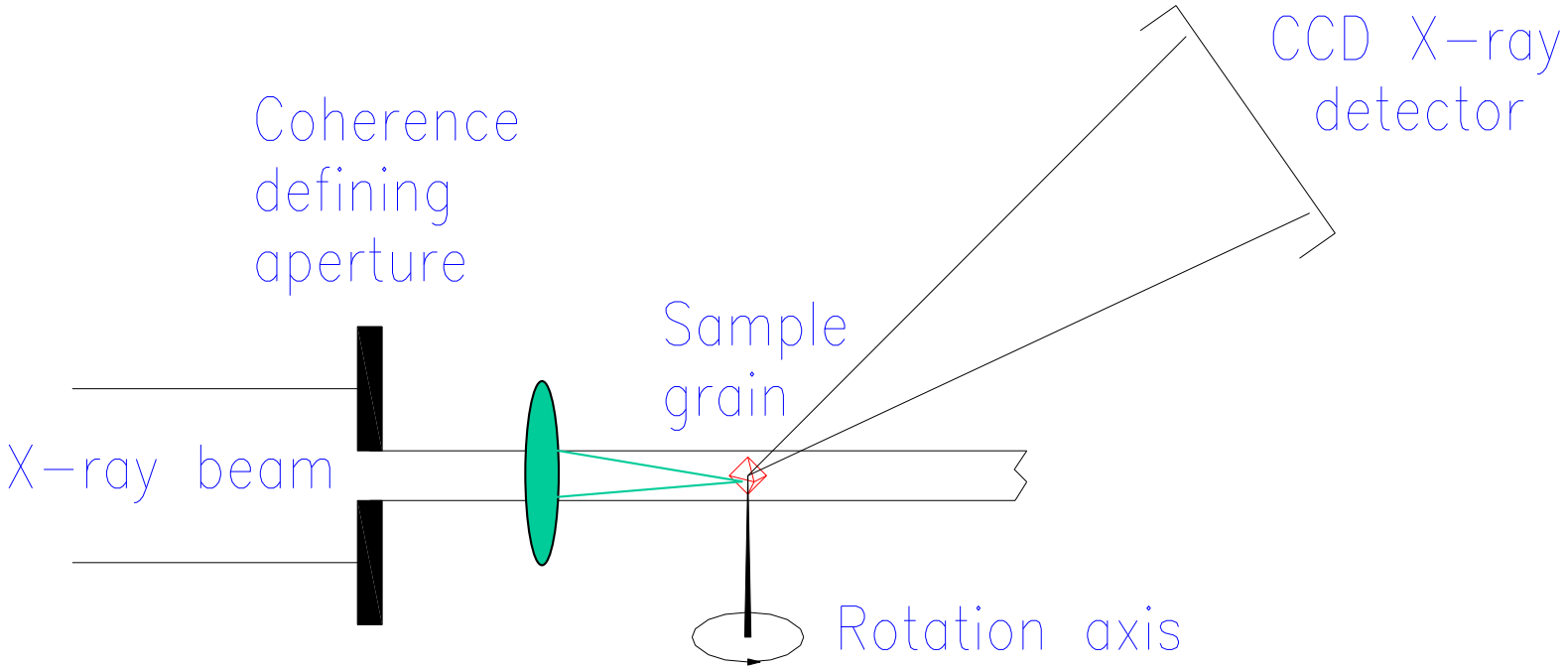
- 500nm Pb crystals grown in-situ in UHV system at APS (Argonne) on a SiO_2 support (vertical).
- 3D coherent X-ray diffraction data is inverted into hemi-spherical cross-sectional slices.

- These images are colored according to the phase of the density, which changes abruptly at an internal boundary.
- This plane is identified as a single deformation fault in the crystal.

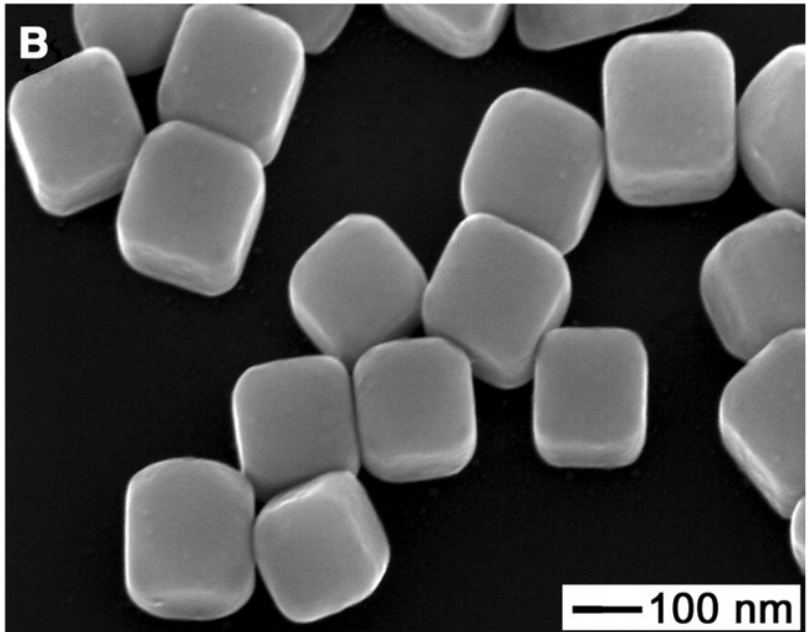

MICRO-CONTROLE
Newport

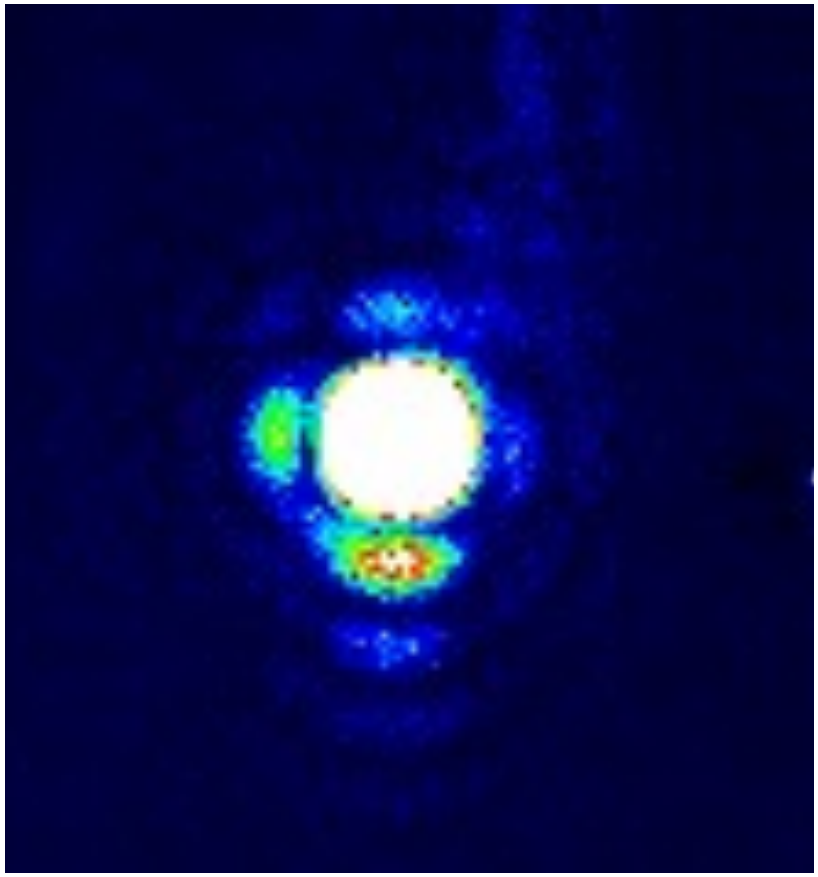

MODEL HSC-1

Lensless X-ray Microscope



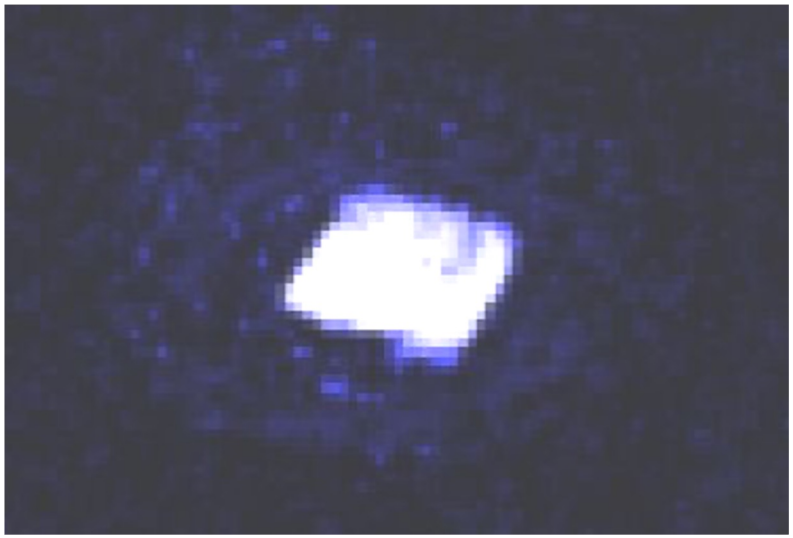
CXD from Silver Nanocubes



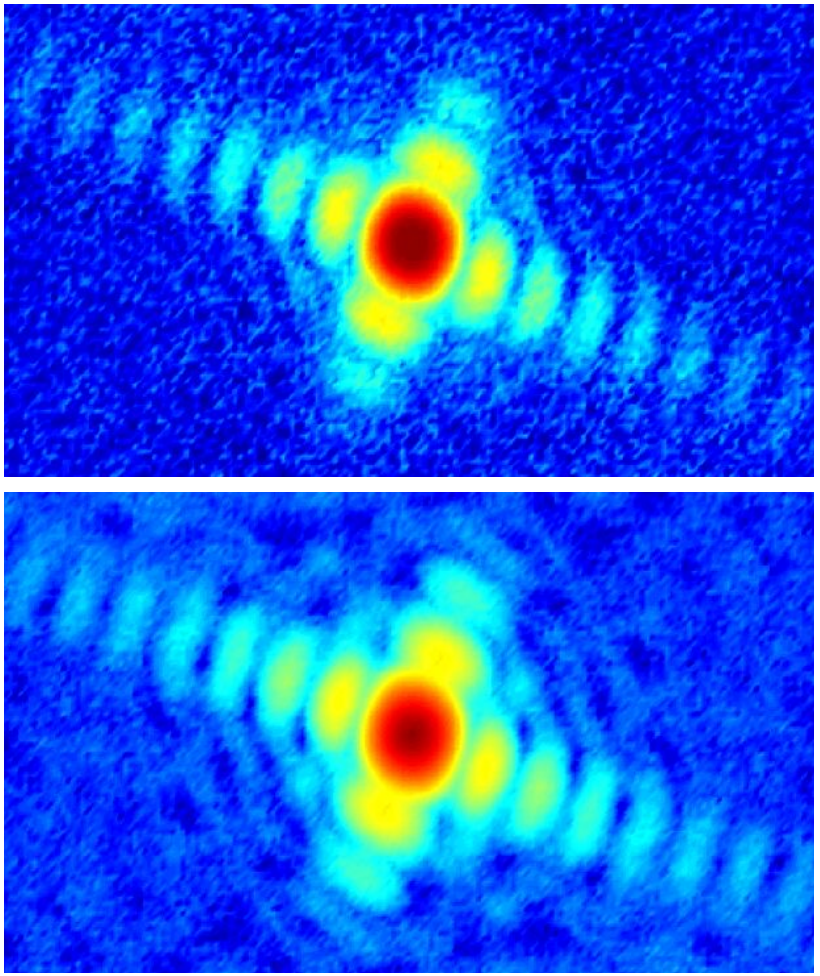


Yugang Sun and Younan Xia,
Science 298 2177 (2003)

Reconstruction of Ag Nanocrystal

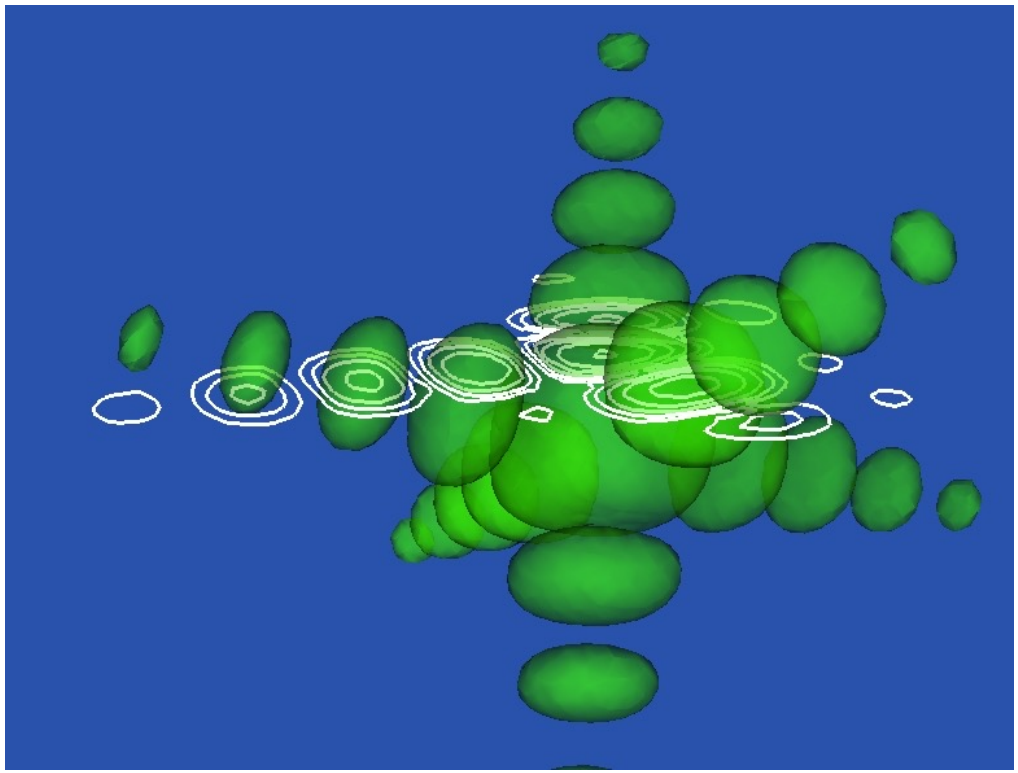


200nm



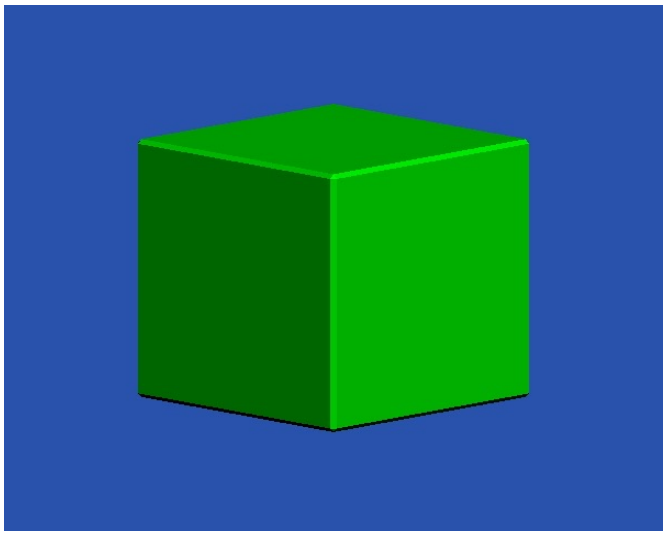
Miscentered Non-Centrosymmetric Reciprocal Space Slice

$$A_q \neq A^*_{\bar{q}}$$

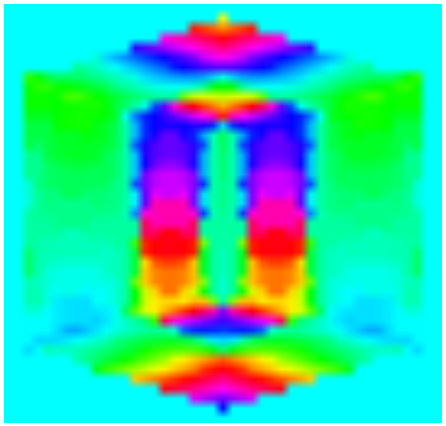


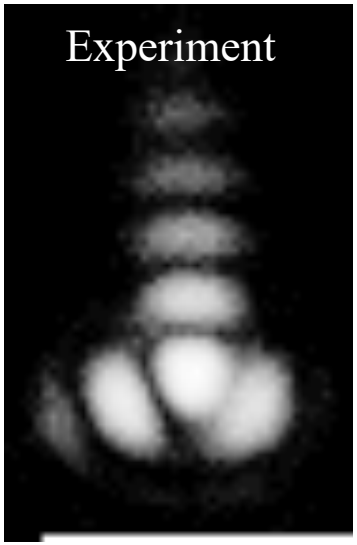
Real-space phase.

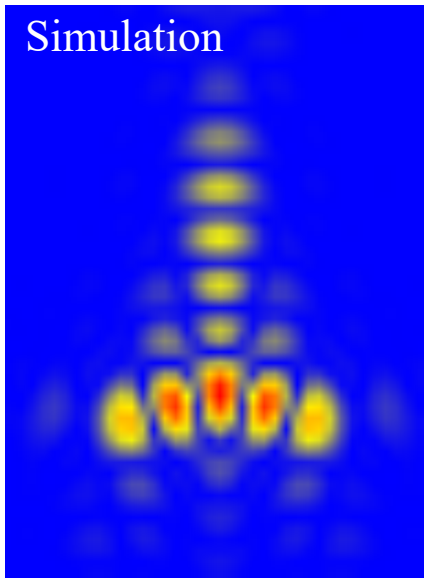
What can we expect to see?



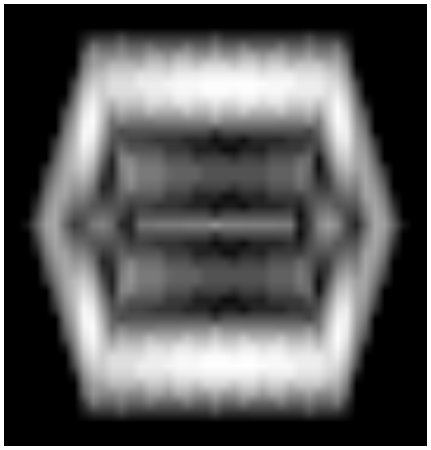
$$A(\bar{q}_o+\bar{q}_z)=\sum_i \sum_z \underbrace{\rho(x,y,z)}_{u_i(x,y)} e^{2\pi i \bar{q}_z\cdot\bar{z}_i} e^{2\pi i(\bar{q}_o)\cdot\bar{r}_i}$$

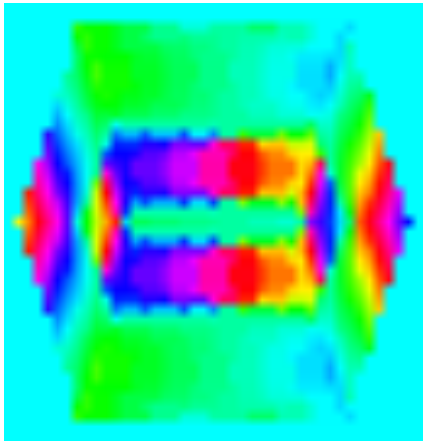


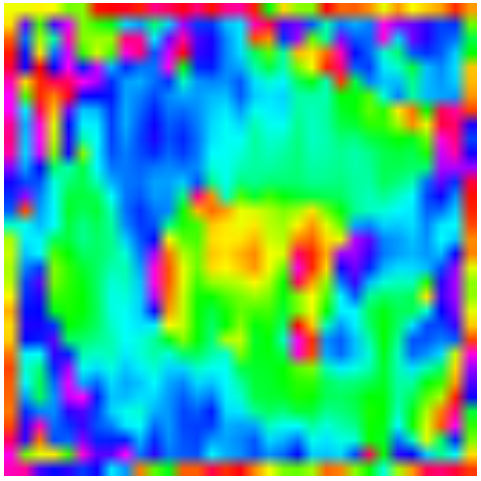


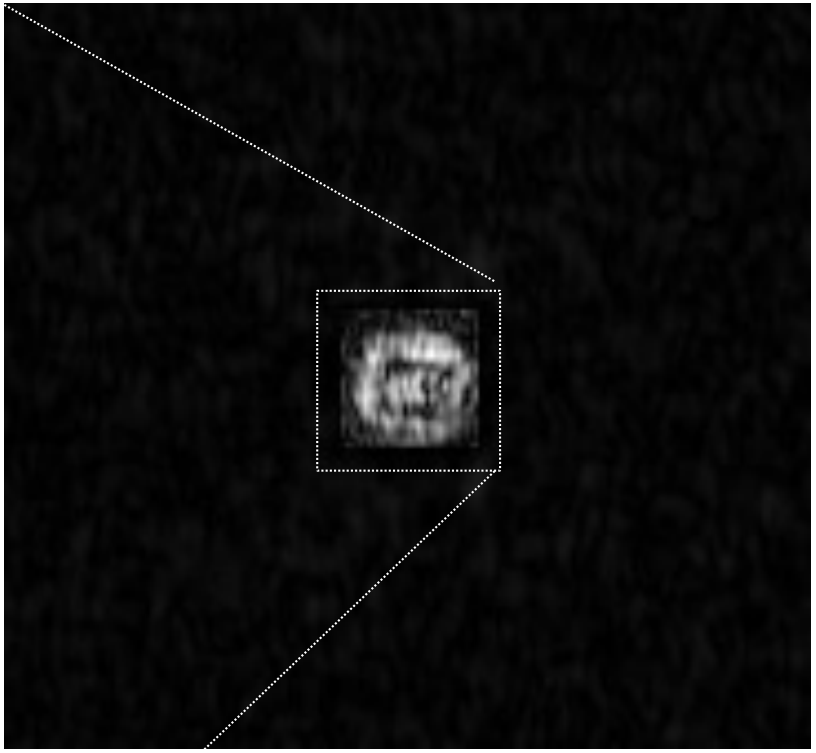


Direct Space Result (1000 Iterations)

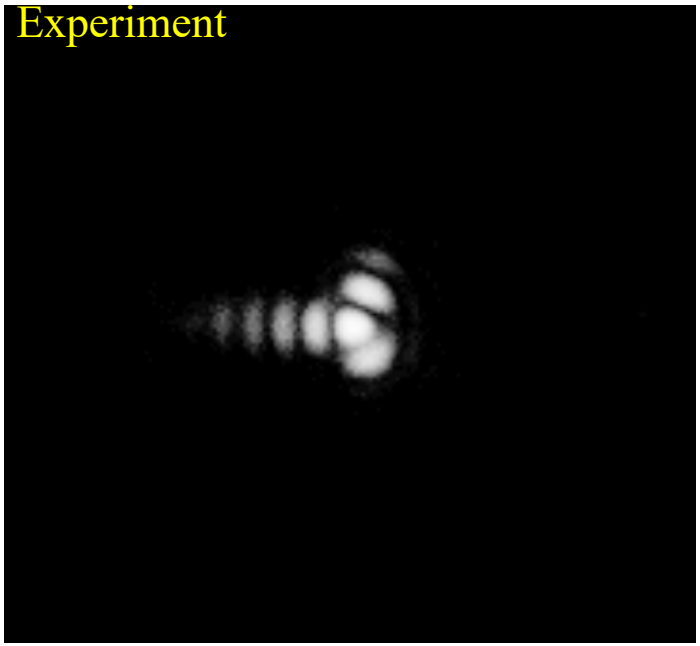


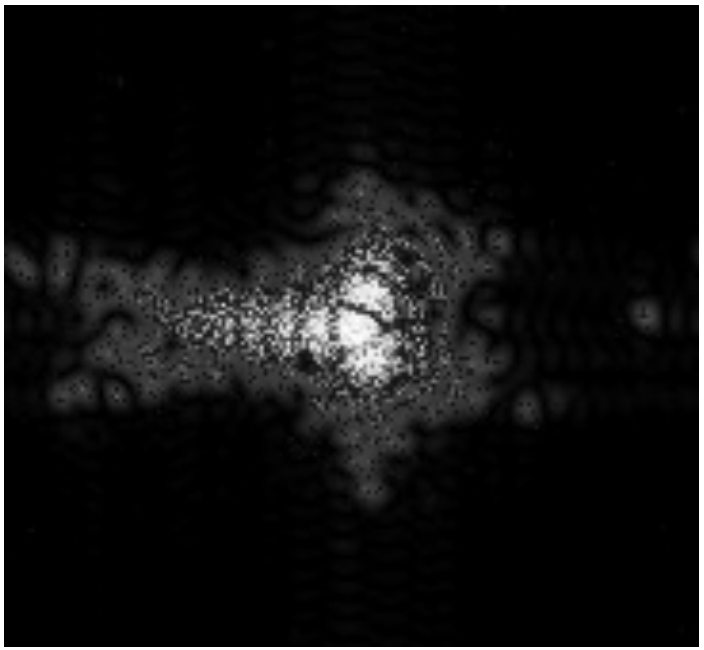


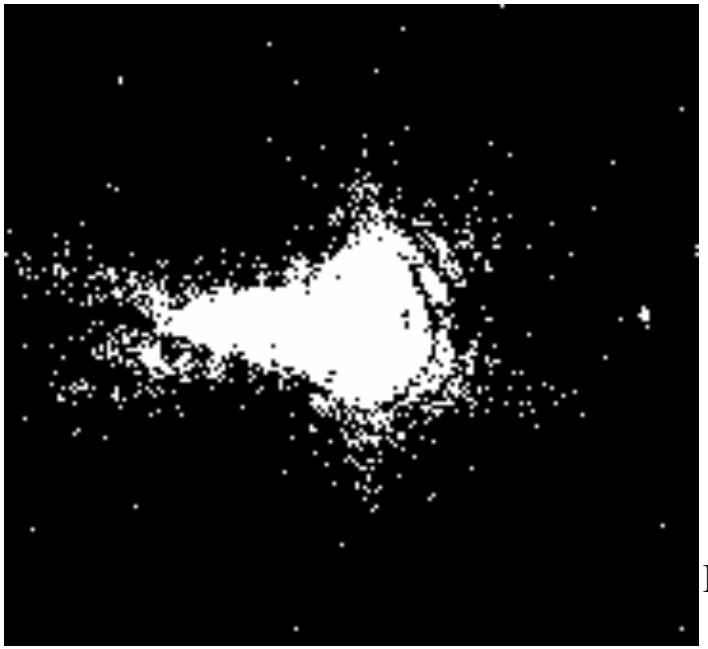


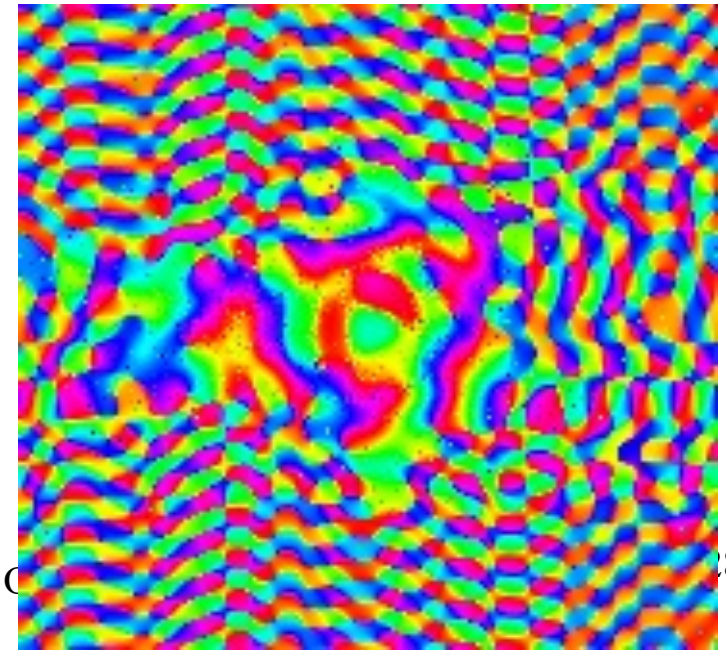


Reciprocal Space Result (1000 iterations)

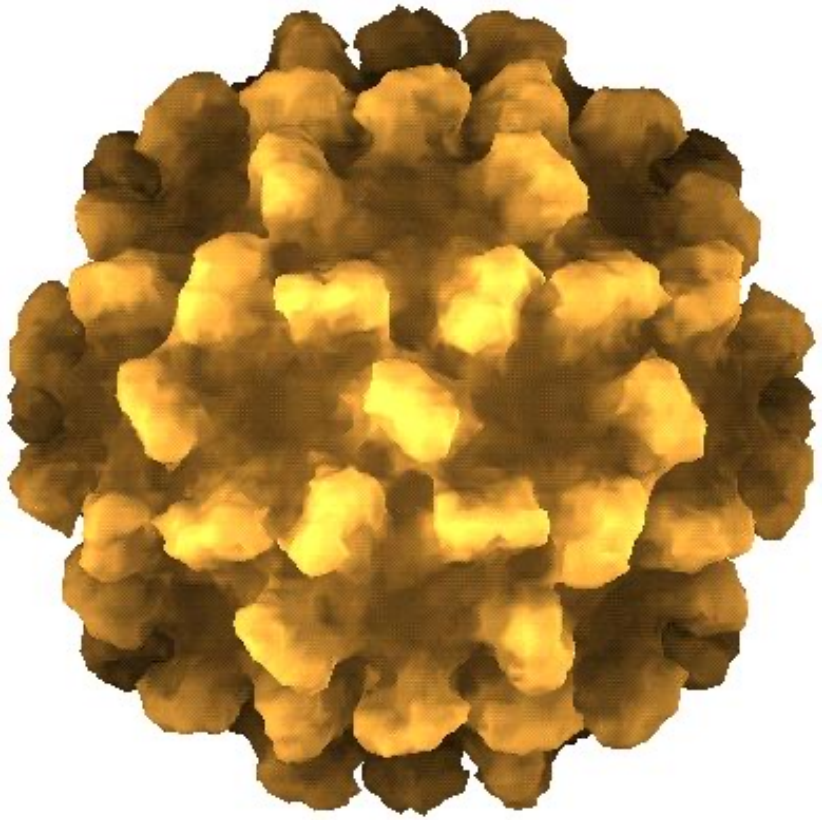








Tomato Bushy Stunt Virus
1980



Conclusions and Outlook

- Inversion of CXD demonstrated
- Internal structure of Au Nanocrystals
- Preservation of coherence upon focussing
- Off-center diffraction gives complex image
- Single molecules one day